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Evolving Metallurgical Behaviors in Plutonium from Self-Irradiation

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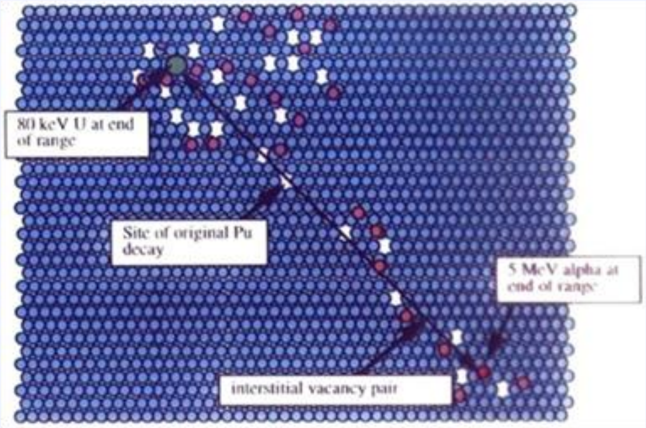
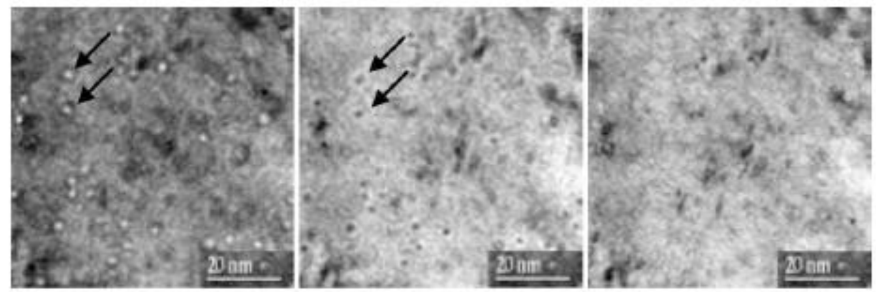
We need to assess the impact of aging on metallurgical properties of plutonium

- Assessment of aging effects in plutonium is needed in developing predictive capabilities to describe the properties of plutonium
  - Alpha decay and the associated recoil of a uranium atom produce damage in the plutonium lattice that accumulate over time
  - We must understand how this damage affects the properties of plutonium alloys as a function of their age
- This work measures the density and mechanical property changes observed from both naturally and accelerated aged plutonium alloys
  - Dilatometry, immersion density, quasi-static tensile and compression

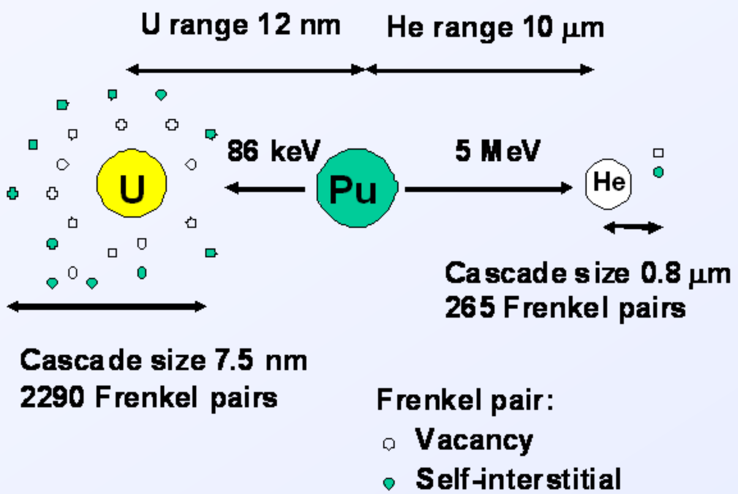


The plutonium alpha-decay leads to aging effects

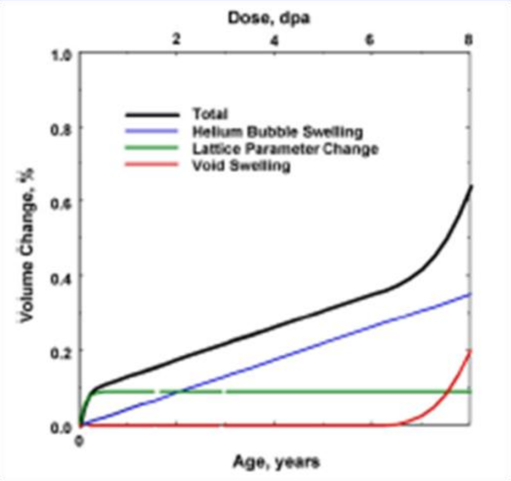
- The alpha-decay in plutonium creates lattice damage, radiogenic helium and actinide daughters
  - The cumulative rate of helium in-growth is about 41 atomic parts per year



A. Schwartz et al., Phil. Mag., v85, p479 (2005)



W. Wolfer et al. Los Alamos Science, v26, p274 (2000)



W. Wolfer et al., J. Alloys Compds., v444-445, p72 (2007)

Helium in-growth is generally known to be the dominant effect during extended aging

We used natural and accelerated aged alloys to measure aging effects on density and mechanical properties

Accelerated Aged Alloy Preparation

- We cannot afford to wait for long periods to measure aging effects in plutonium
- Accelerated the aging process by doping with ^{238}Pu
  - Damage process and defect types are identical to reference materials
- Pu alloys are enriched with approximately 7.5 wt% ^{238}Pu
  - Acceleration rate based on the difference in the activity of the spiked alloy to that of the reference alloy – Initial rate is roughly 18 fold increase
  - This rate will decrease as the material ages due primarily to decreasing concentration of ^{238}Pu in the spiked alloy

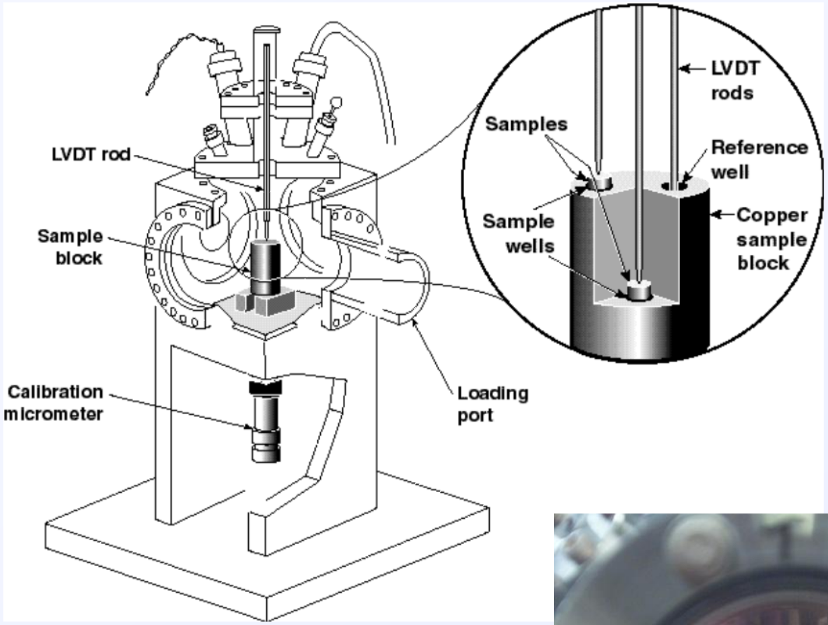
Density Measurements

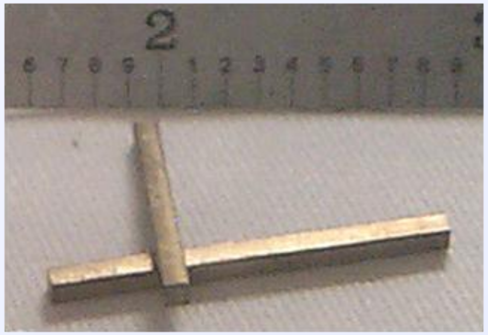
- Dilatometry
- Immersion density
- Aged natural alloys (PA), Reference alloys (RA), and Accelerated aged alloys (AA)

Mechanical Tests

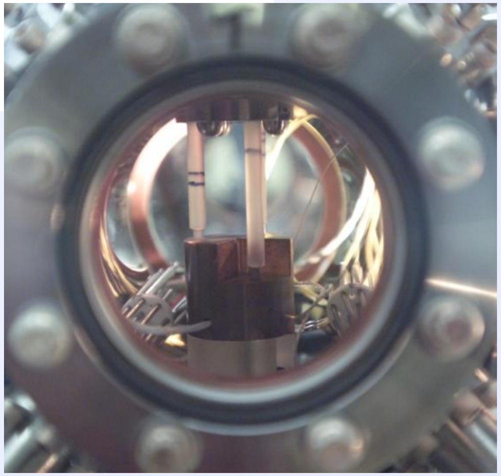
- Tensile at RT (quasi-static)
- Compression at RT (quasi-static)
- Aged natural alloys (PA), Reference alloys (RA), and Accelerated aged alloys (AA)

Dilatometry System is Designed for Maximum Sensitivity and Stability





Dilatometry samples



Dilatometry system

Design based on Linear Variable Displacement Transducer (LVDT)

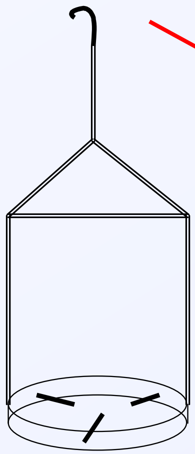
Two different lengths (2 and 3 cm) are used to differentiate between surface oxidation and volumetric swelling

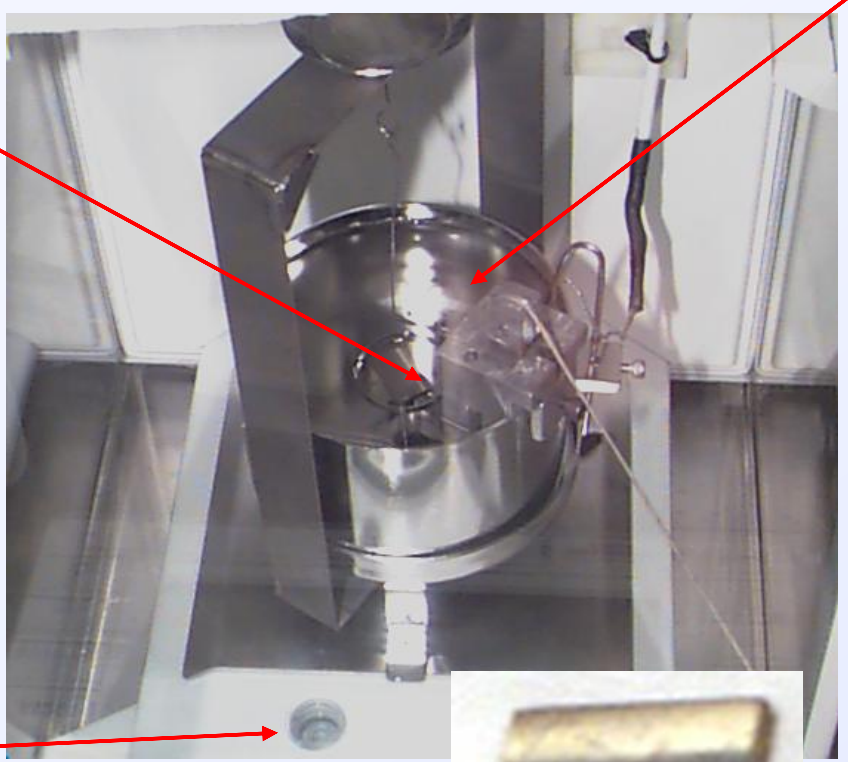
Third well contains zero-dur as reference material

Stability over 120 days is better than
- ± 0.10 μm for the LVDTs
- ± 0.03°C for the Cu block

Immersion Density Technique is Used to Measure Density of both Reference and Spiked Pu Alloys

Sample holder





Balance

FC-43 Inert fluorochemical as the displacement media

1.9423 g/cc, -0.0021 g/cc per °C

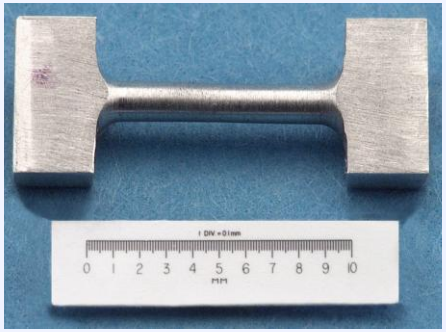
Prior to use, the system is calibrated using NIST glass (SRM 1827A)

A correction needs to be applied to the measured density to compensate for the heat generated by ^{238}Pu

Test equipment is based on immersion density equipment design by H. A. Bowman, et al., *J. of Res. Nat. Bur. Stand.*, 71C (3), 179-198 (1967)

Tensile Test Equipment and procedural methodology to meet QA compliance

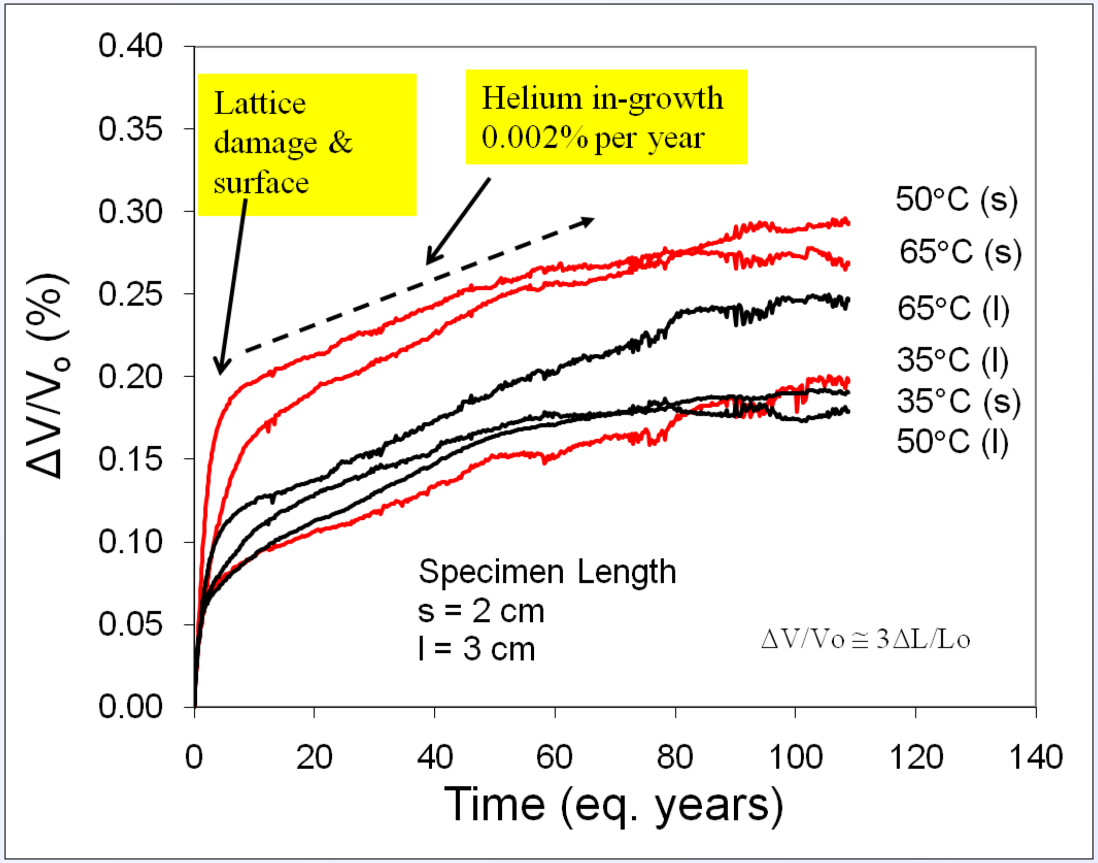
- INSTRON Model 4444 Single Column Testing System. Cap. 2kN (200kg, 450 lb)
- Series IX®Software for Windows to run tests, analyze results and store data
- Practice runs upon a comprehensive operating procedure using surrogate samples Al(1100)



Tensile part
Gauge diameter
0.060 ± 0.001 inches
Gauge length
0.316 ± 0.001 inches

Compression part
Diameter
0.100 ± 0.001 inches
Length
0.300 ± 0.001 inches

Dilatometry on Spiked Pu alloys with ~0.6 wt. % Ga shows volume swelling with age



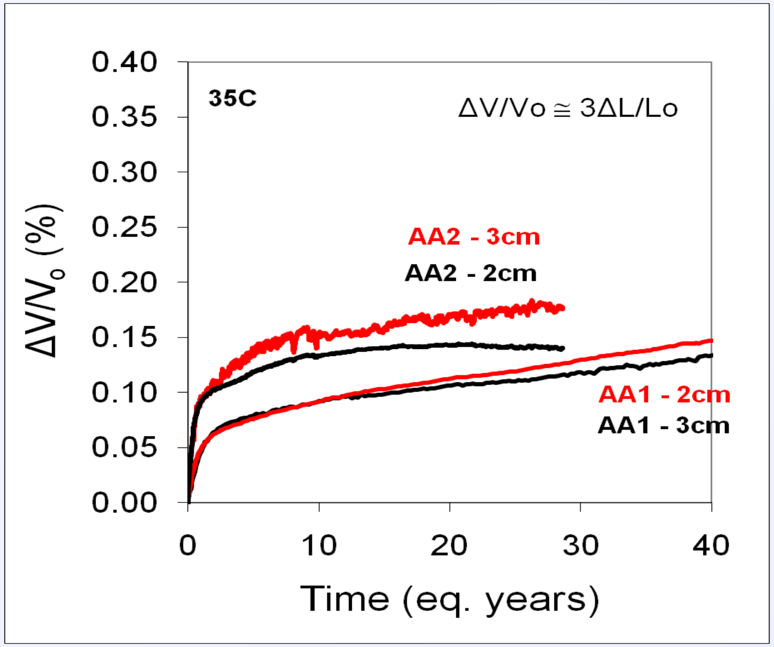
- **Sample temperatures maintained at 35, 50 & 65C under helium atmosphere**
- **Small fluctuations in data caused by external environments affecting data acquisition systems**
  - **Outside temperatures**
  - **External vibrations**

Equilibrium swelling does not exceed 0.002%/year

The rate of volume change decreased after ~60 eq. years

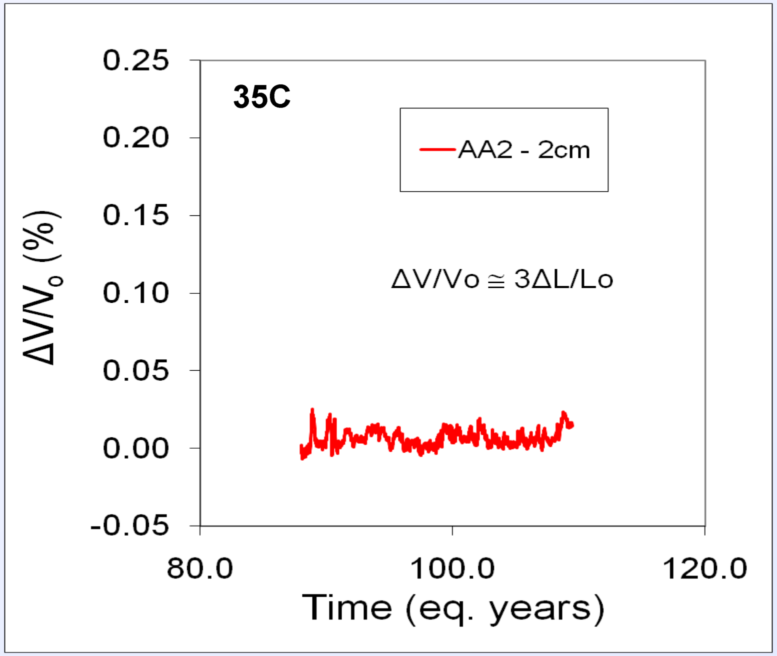
Spiked Pu alloys with ~1 wt. % Ga at 35°C show volume swelling with age

Spiked Pu alloys with ~1wt. % Ga (AA2)



- AA2 alloys were annealed at 150°C for 30 minutes to remove accumulated lattice damage (eq. age of 86 years)
- Initial transient indicates larger change compared to AA1 alloys (~0.07% at 3 eq. years)

Spiked Pu alloys with ~1wt. % Ga (AA2)



- AA2 alloy aged to eq. ages of 86 years does not show significant volume changes
  - Similar to aged AA1 alloys

Dilatometry on both spiked and reference alloys shows volume swelling due to lattice damage and helium in-growth

Data Analysis and Findings

- The known contributors to the inverse exponential-type and linear volume swelling are changes in lattice parameters and the build-up of helium, respectively.
    - The curves at 35°C are quite accurately represented by the combination of the exponential and linear growth

$$\Delta V/Vo = A\,[1-\exp(-Bt)] + Ct$$

    - While the curves at 50 and 65°C are influenced by possible surface oxidation during the initial stage of swelling, helium in-growth dominates the linear growth

    - Helium-to-vacancy ratio can be approximated with the linear swelling rate and the helium-induced expansion relationship

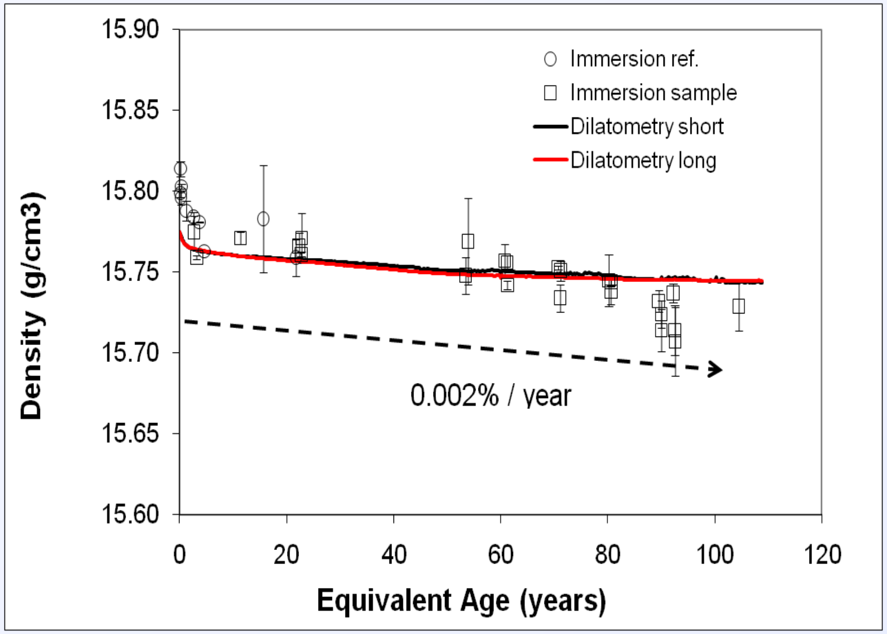
$$C = (\Delta V/Vo)He/t \cong [He]/(ratio)$$

[He] = 41.1 appm per year & t is time

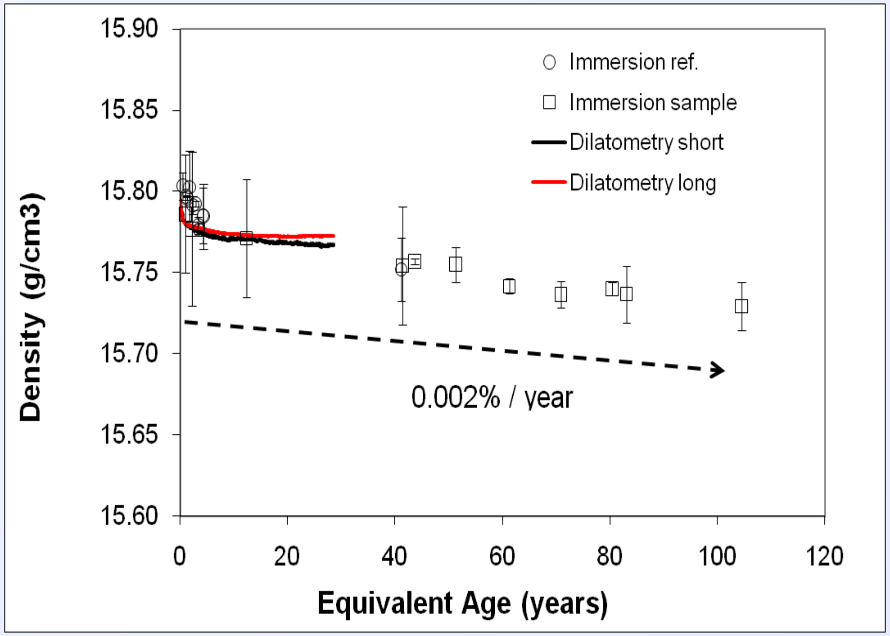
- At less than 60 eq. years, the average He/Vacancy ratio is 2.5±0.6 - AA1
- At greater than 60 eq. years, the rate of volume change decreases - AA1
    - Similar observation made with aged AA2

Immersion density of spiked, reference, and naturally aged Pu alloys shows gradual decrease with age

~0.6 wt. % Ga alloys (AA1 and RA1)



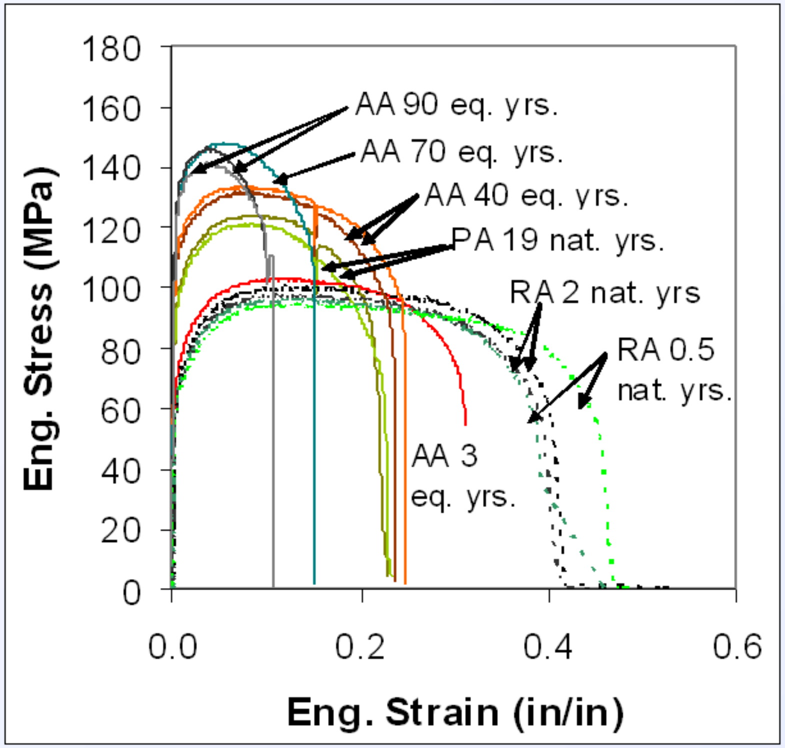
~1 wt. % Ga alloys (AA2 and RA2)



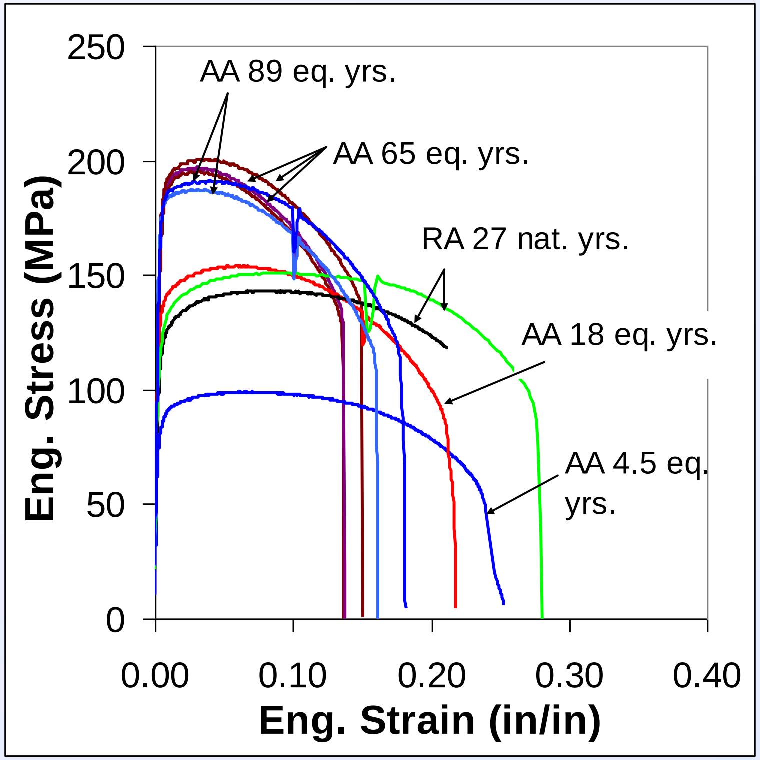
- **Pu alloys show gradual decrease in immersion density with age without signs of void swelling**
- **Density samples are stored in a helium-flushed incubator maintained at 50°C between measurements**

Tensile tests on Pu alloys at ambient temperature and pressure shows increase in strength due to the aging process

~0.6 wt. % Ga doped Pu alloys



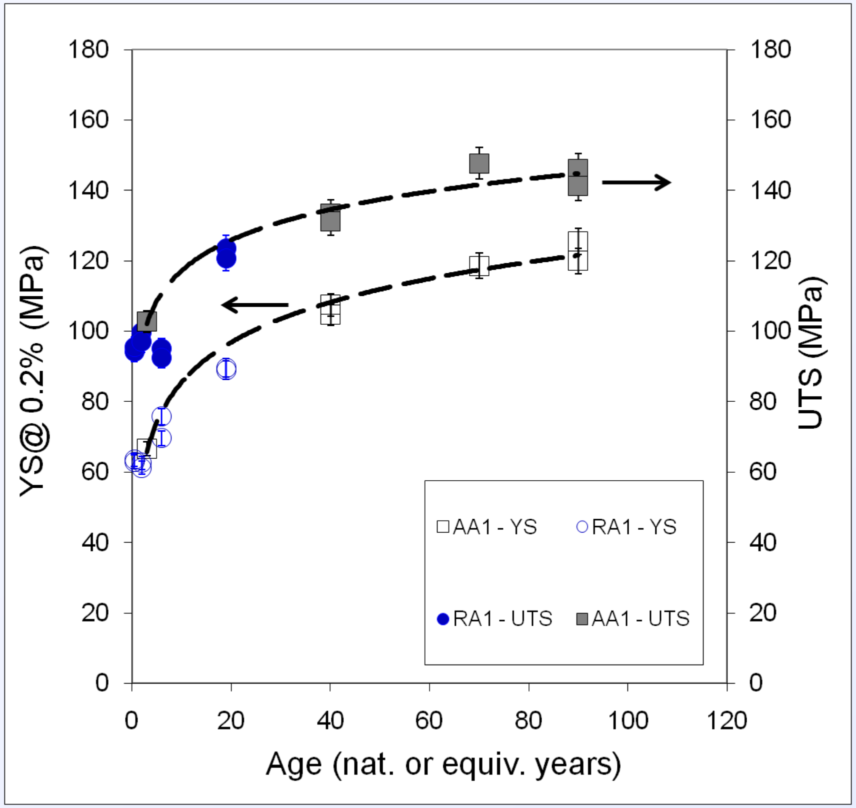
~1 wt. % Ga doped Pu alloys



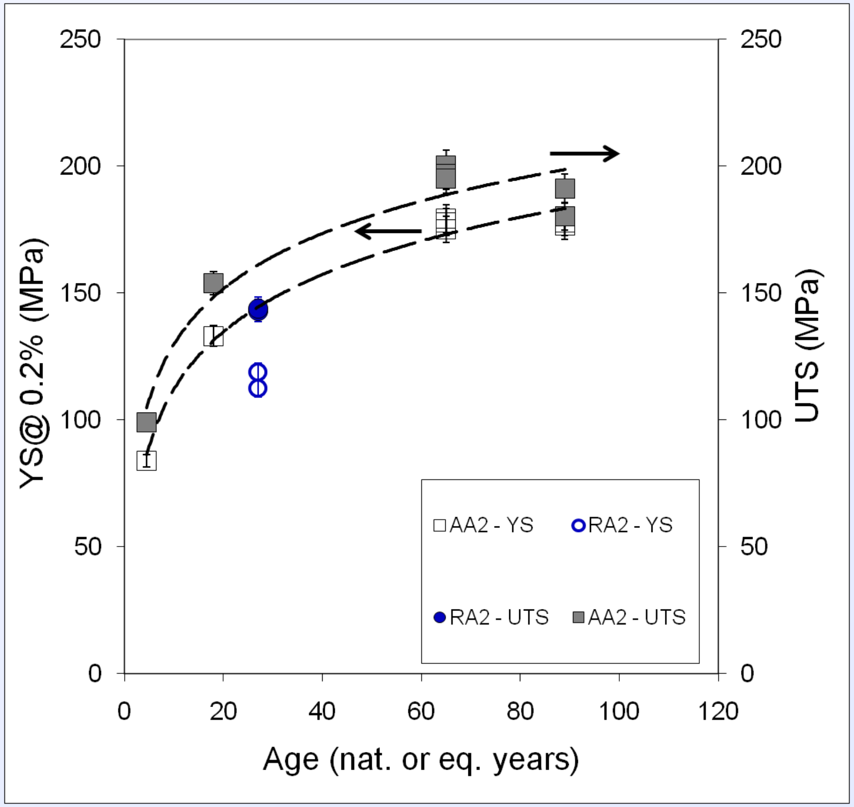
- Both alloys show increase in the yield strength (YS) and the ultimate tensile strength (UTS) with age.
- Possible saturation of YS and UTS after 70 eq. years of aging
- Slight increase in Young's Modulus, from ~40 GPa to ~45 GPa
- The ductility decreases with age

Tensile tests show increasing yield strength and ultimate tensile strength as plutonium ages

~0.6 wt. % Ga doped Pu alloys



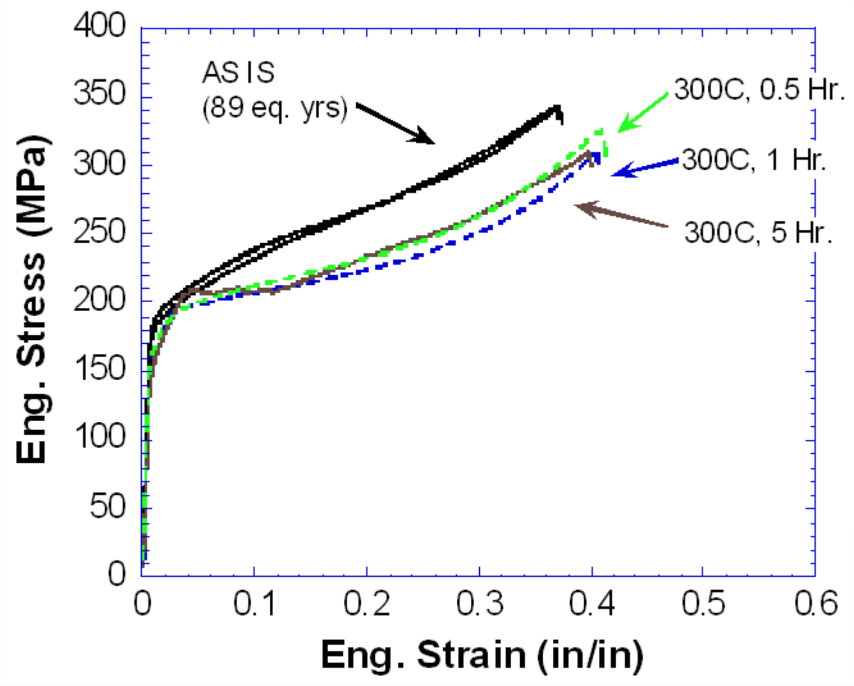
~1 wt. % Ga doped Pu alloys



Tests are showing saturation in YS and UTS between 70 and 90 eq. yrs. of aging

Helium in-growth contribution to static mechanical property can be studied by annealing

Static compression tests on enriched Pu alloys (AA2)



- **The yield strength from aged (~89 eq. yrs.) plutonium alloy is 172 MPa**
  - **Combined contributions from intrinsic, lattice damage, and He in-growth components**
  - **Intrinsic yield strength is ~80 MPa**
- **The annealed yield strength is ~140 MPa**
  - **Reduction of ~30 Mpa due to annealing out the accumulated lattice damage**
- **The in-growth of helium contributes ~60 MPa for this alloy aged to 89 equiv. years**
  - **Saturation?**

- **Annealing experiments can be applied to improve our understanding of aging effects to metallurgical properties of plutonium**

Metallurgical properties of spiked and naturally aged alloys evolve by aging process

- Dilatometry and immersion density show decrease in densities of ^{238}Pu spiked alloys (AA), reference (RA), and aged stockpile (PA) alloys from the aging process
  - Average He/vacant site ratio is 2.5 to 60 equivalent years, maybe changing at time greater than 60 eq. yrs.
- Void swelling is not yet observed
- The change in density calculated from the dilatometry corresponds well to the immersion density of spiked and naturally aged Pu alloys
- Static tensile and compression tests show increase in strength and decrease in ductility with age
  - Both lattice damage and helium in-growth contributes to changes in mechanical properties
    - Possible saturation effects after ~60 eq. years of aging
  - Contribution of helium in-growth to mechanical strength can be studied by annealing experiments

Plan to continue testing both spiked and naturally aged alloys to assess the aging effects in plutonium alloys

- We need further work to determine the cause for change in $\Delta V/V_o$ after aging 60 equivalent years
  - Helium bubble size, extended defects (more than simple Frenkel pairs?), transuranic in-growth, etc
- There appears to be minimal change in YS and UTS after 60 eq. yrs. of aging
  - Possible saturation effect?
- Additional annealing tests on aged samples to improve aging effects in Pu alloys
- Additional efforts in metallography and microscopy started to support density and mechanical tests